

LA-UR- 04- 4187

Approved for public release;
distribution is unlimited.

Title:

**SAFEGUARDABILITY OF ADVANCED SPENT FUEL
CONDITIONING PROCESS**

Author(s):

**T. K. Li, S. Y. Lee, T. L. Burr, K. E. Thomas, P. A. Russo,
H. O. Menlove, H. D. Kim, W. I. Ko, S. W. Park, and H. S.
Park**

Submitted to:

**45th Annual INMM Meeting
Orlando, FL
July 18-22, 2004
(FULL PAPER)**



Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.



SAFEGUARDABILITY OF ADVANCED SPENT FUEL CONDITIONING PROCESS

T. K. Li, S. Y. Lee, T. Burr, K. Thomas, P. Russo, H. Menlove
Los Alamos National Laboratory
P.O. Box 1663, Los Alamos, NM 87545, USA

H. D. Kim, W. I. Ko, S. W. Park, and H. S. Park
Korea Atomic Energy Research Institute
P.O. Box 105, Yusong, Taejon 305-600, KOREA

ABSTRACT

The Advanced Spent Fuel Conditioning Process (ACP) is an electro-metallurgical treatment technique to convert oxide-type spent nuclear fuel into a metallic form. The Korea Atomic Energy Research Institute (KAERI) has been developing this technology since 1977 for the purpose of spent fuel management and is planning to perform a lab-scale demonstration in 2006. By using of this technology, a significant reduction of the volume and heat load of spent fuel is expected, which would lighten the burden of final disposal in terms of disposal size, safety and economics. In the framework of collaboration agreement to develop the safeguards system for the ACP, a joint study on the safeguardability of the ACP technology has been performed by the Los Alamos National Laboratory (LANL) and the KAERI since 2002. In this study, the safeguardability of the ACP technology was examined for the pilot-scale facility. The process and material flows were conceptually designed, and the uncertainties in material accounting were estimated with international target values.

INTRODUCTION

The question of “how to manage the spent fuel discharged from reactors” is a key factor for the sustainability of nuclear energy. Approximately 6,000 metric tons of spent nuclear fuel from reactor operation has been accumulated in South Korea with expectation of more than 30,000 metric tons, three times the present storage capacity by the end of 2040 [1]. The ACP technology was designed to address these challenges, through the development of fuel cycle technologies coupled with advanced safeguards that will meet domestic and international nuclear materials management needs. The Electrolytic Reduction (ER) technology developed recently by KAERI is being expected as a more economic, efficient and proliferation resistant concept for the conditioning of spent fuel. The electrolytic lithium reduction process uses molten LiCl to reduce the oxide components of the spent nuclear fuel, yielding the corresponding metals and Li_2O . The goals of the ACP are to recover more than 99% of the actinide elements and to minimize the volume and heat load. The metallic product will be collected separately and held in interim storage facility until its ultimate disposition is decided.

This paper summarizes the preliminary results of collaboration between the LANL and the KAERI for the assessment of safeguardability on ACP. The sub-processes and material flow of the pilot scale ACP facility were designed, and then their material balance area (MBA) and key measurement

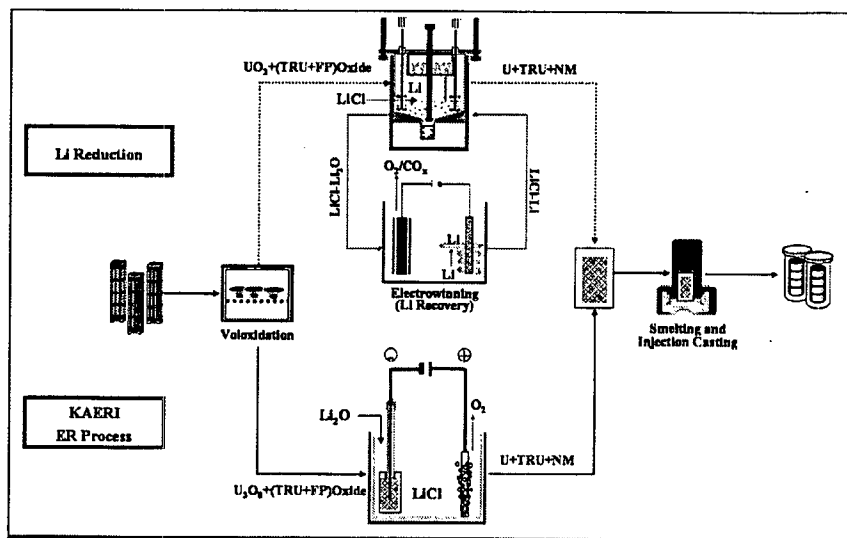


Fig. 1. Flow Diagram of Lithium Reduction Process.

point (KMP) were defined based on diversion scenario analysis. The uncertainties in material accounting were also estimated with international target values for the uncertainty of measurement methods.

MAIN PROCESS CONCEPT

The ACP technology is based on the pyro-chemical process that was designed in the 1960s and 1970s. The reference concept consists of six major sub-processes that are illustrated in Fig. 1. They include;

- dismantling the fuel assemblies, cutting fuel rods, and removal of the cladding,
- thermal oxidation of UO_2 to U_3O_8 ,
- reduction of the oxide fuel into metals, using a suitable reductant in a molten salt,
- recovering of the reductant metal by electrolysis of its oxide to allow recycling it and to minimize the waste generation,
- smelting of metalized fuel, and
- casting of metalized fuel in a form that is suitable for interim storage and deposition.

In the reference lithium reduction process, the oxide fuel elements are chopped into segments and are voloxidized, and the resultant oxide powder is loaded into a porous magnesia basket. The baskets are charged into a reduction vessel, where the fuel is reduced with lithium dissolved in molten LiCl at 650°C . Some fission products with high heat load such as cesium and strontium are dissolved in lithium chloride molten salt, and separated from the spent fuel product [2].

Recently, a modified concept of Li reduction had been proposed by KAERI to simplify the process and to increase the proliferation resistance of the process based on the reference technology. In the electrolytic reduction (ER) concept, the lithium electro-winning step is conducted in the uranium

oxide cathode simultaneously and there is no step for salt recovering. In this concept, as shown in Fig. 1, the lithium is recovered electrolytically at the uranium oxide cathode and this lithium reduces oxides in spent fuel to metal. Consequently, lithium recovery process is no longer needed in this concept and the possibility of separation of actinides is inherently ruled out.

INHERENT ATTRIBUTES

The success of the ACP will depend on a number of factors. One of key factor would be proliferation resistance, and it would be judged by the manner in which it addresses the issue of proliferation. The existing “open” or “once through” LWR fuel cycle is relatively proliferation resistant compared to closed cycles. As long as the fuel assemblies remain intact, the safeguards approach is straight forward, but may be resource intensive for large numbers of assemblies. If the assembly is broken down, proliferation risk increases from loss of identity of the assembly and the huge numbers of fuel pins that must be tracked. Breaking open pins to process the fuel introduces significant proliferation risk from bulk-handling operations. Any closed fuel cycle is likely to present an increase in proliferation concerns, and bulk-handling operations are a perfect diversion location because of the availability of the material and reliance upon materials accounting for detection. Material accounting must be the most sophisticated for these processes, and even then it may not be able to detect diversion for large processing plants [3].

Not all nuclear facilities, however, are equally susceptible to proliferation purposes nor are they all equally easy to safeguard. Intrinsic factors influence both the attractiveness of materials/facilities to proliferators and their safeguardability. There would be several inherent attributes of the ACP process that make this fuel cycle unattractive for diversion when compared with conventional fuel reprocessing and plutonium recycling.

- The processes used for the ACP do not produce a pure or partially pure plutonium product.

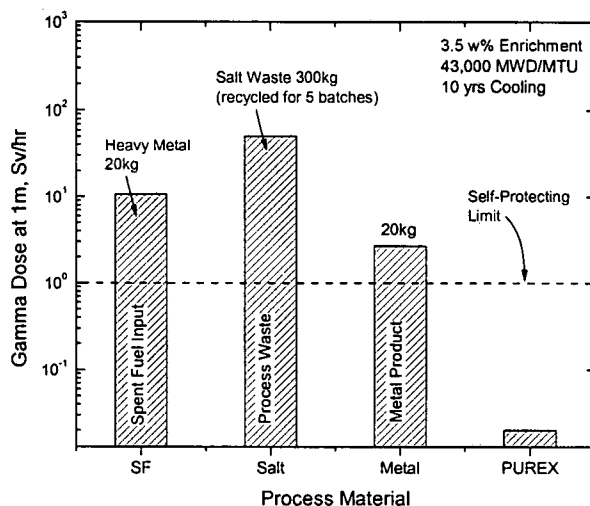


Fig. 2. Self-Protection by Fission Products of ACP Materials.

Because of the chemistry of the ER process, no fissile material can be separated in pure form. Plutonium is co-deposited together with minor actinides and some fission products [4]. Therefore, the material requires further chemical reprocessing to separate pure fissile elements. This results in longer warning times and requires development of a technology standard in order to obtain material suitable for weapons purposes.

- The decay heat and radioactivity of the ACP product (metallic form of the spent fuel) are about 25% of those of the initial spent-fuel feed to the ACP. Nevertheless, as shown in Fig. 2, the presence of some fission products leads to a high dose rate of radiation arising from the metal product. Furthermore, some of the processes used in the ACP require high-temperature furnace operations under controlled atmospheres even in the heavily shielded hot cell. Unauthorized penetrations into the processing cell to divert small quantities of materials using a clandestine material stream on a protracted basis would be extremely difficult and readily detectable.
- The reconstitution options require a highly remote operation in canyons of highly shielded cells. It is difficult to gain undetected access to these cells to modify hardware or install new processes. The complexity of these operations with highly radioactive materials precludes manual operation. The process must be highly automated with inherent abilities to track and log in-cell operations included in the design. Incorporation of this information into a safeguards system will, therefore, not interfere with plant operations.

These inherent features of ACP may be concordant with the PIPEX concept as was proposed during the INFCE [5]. Similar observations have already been made for the IFR fuel cycle, where the material is considered as "self-protecting" for the reasons mentioned above [6]. Nevertheless, it might be mentioned that the self-protecting is not valid when the State is the adversary.

MATERIAL CONTROL & ACCOUNTABILITY

A pilot scale facility with a capacity of 30 MTHM/year was designed in this study to analyze the safeguardability of the ACP. The facility stands alone physically (operationally), and is administratively isolated from reactors and interim spent-fuel storage facilities. The main process of the facility is the ER concept, which has no need of the Lithium recovery system. The facility availability is assumed 60%, which is equivalent to 219 full operating calendar days per year. The process consists mainly of three parts: spent fuel handling area (spent fuel disassembling and rod extraction), main hot cell (decladding, reduction, smelting, casting, etc.), and U-metal handling area (loading metal rods into storage cask and temporary storage). The reference spent fuel used in the facility is Korean Yong-Gwang Unit 1&2 PWR's standard 17×17 assemblies with a minimum 10 years of cooling time after 45,000 MWd/MTU of final burnup.

Material Accounting System

Lacking specific design information for the ACP facility, the features such as the MBA definition, material flow pattern, KMPs, and inventories on material balance closing were designed for the conceptual facility. Many assumptions necessary to calculate the detection sensitivity of the materials accounting system were also made. The ACP fuel conditioning facility was designed to be composed of two MBAs [7]. The operations of MBA-A are based on individual item counts because the composition is not varied and items are only broken into other discrete items. Therefore, the

material accountancy in the MBA-A is similar to that in any storage area.

Figure 3 identifies MBA-B boundaries, KMPs, and locations of inventories at material balance closing. It is assumed that the facility closes material balances once every 3 months or once after 54 days of operation. It is also assumed for this analysis that the present IAEA detection goals for spent LWR fuels would be applied to materials within the ACP facility. Nuclear material contents for material balance were calculated based on the reference fuel and the material contents. In MBA-B, the facility operator does material accounting based on some declared values for feed materials; destructive chemical analyses for mixed oxides and metal ingots; and NDA measurements for U-metals, recyclable scraps, and disposable waste streams. Isotopic analysis for ACP materials with respect to mass distribution, total dose rate, and neutron production rate recommends that a curium-monitoring method could be available if the amount of Pu relative to Cm is verified continuously at all stage of the process [8].

Since the size, shape and chemical form of nuclear material would be changed in the ACP, a more sophisticated material accountancy method is required. Two types of material balance concepts are employed: batch closeout, which is the inventory difference for a single process, and material

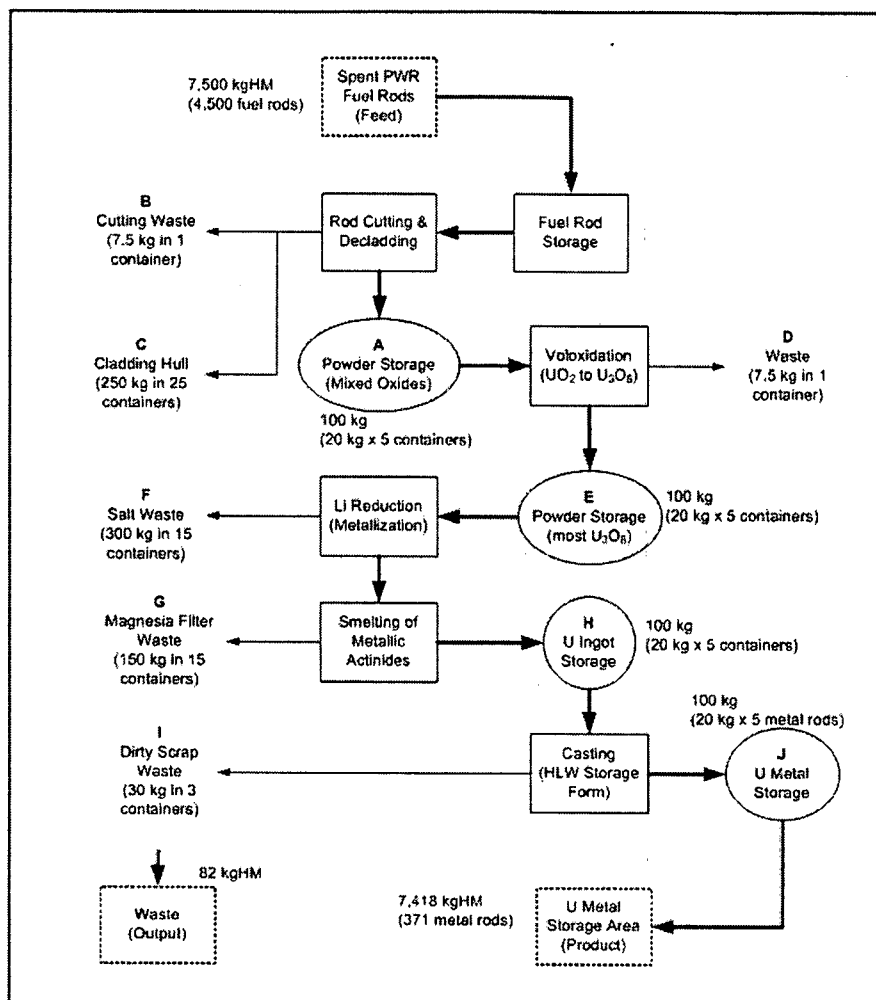


Fig. 3. Material Inventory at MBA-B of Conceptual ACP Facility.

Table 1. Characteristics of the ACP Strata for Material Accounting at ACP Facility.

Stratum	KMP	Material Form	Total Element (kg)	Total Pu (kg)	Accounting Method
1	1	Spent Fuel Feed (most UO_2)	7500.00	88.875	DA + Weigh
2	2	U-Metal Product (TRU+MA)	7440.00	88.164	NDA
3	3	Waste Output	60.00	0.711	NDA
4,14	A	Mixed Oxides Storage (most UO_2)	100.00	1.185	DA + Weigh
5,15	B	Cutting Waste (for 1 MB period)	7.50	0.089	NDA
6,16	C	Cladding Hull Materials (for 5 batches)	0.50	0.006	NDA
7,17	D	Disposable Waste & Dirty Power Residues (accumulated for 1 MB period, most U_3O_8)	7.50	0.089	NDA
8,18	E	Mixed Oxides (most U_3O_8)	100.00	1.185	NDA
9,19	F	Salt Waste (accumulated for 5 batches)	1.00	0.012	NDA
10,20	G	Magnesia Filter Waste(for 1 MB period)	7.50	0.089	NDA
11,21	H	Uranium Ingot (for batch closeout)	100.00	1.185	DA + Weight
12,22	I	Dirty Metal Scrap (for 1 MB period)	15.00	0.178	NDA
13,23	J	Uranium Metal Rods(for batch closeout)	100.00	1.185	NDA

accountancy, which is an inventory difference in a specified time interval over several critical zones. The batch closeouts have two different steps based on available information. First, a mass balance is performed based on the total weights of the materials that enter and leave a piece of equipment during a batch. This balance must meet a specified accuracy or operations are halted to investigate possible sources of error. The check provides the assurance that operations proceeded as planned and the inventory difference from the measured weights lies within expected limits. After analytical chemistry results are received, a second batch closeout is performed, which checks expected and measured compositions. The expected masses and compositions of new items are based on operational models and prior experience. This two-step closeout provides the best data for every item in the MBA-B. This system also provides a model of discrete accountable items distributed in space and time and constitutes a complete historical record [9].

IAEA verification would employ attributes and variables measurements, preferably NDA measurements. The facility closes material balances once every three months and plans to have the IAEA inspections coincide with this schedule for plant shutdown, cleanout, and material balance closing. The large inventories of feed materials and products (MBA-A and KMP-J) are maintained as "items" for inventory purposes and are stored in separate storage locations. The cleanout operation before material balance closing recovers almost the entire residual process holdup, and therefore, inventory of plutonium as process holdup is negligible.

Uncertainty Assessment

The Limit of Error in MUF (LEMUF) value was determined based on a hypothetical operating scenario to investigate if the ACP facility would meet the detection goal of IAEA. Because of insufficient detailed information on ACP facility to treat these issues at this time, assumptions regarding measurement procedures on the part of the facility and inspectorate were introduced.

Inventory for the bulk-handing area was assumed as shown in Fig. 3. The characteristics of 23 strata identified in ACP facility are summarized in Table 1. There are one bulk measurement method, three material type determinations, and four analytical methods in ACP. It is important from the

Table 2. Measurement Uncertainties for Material Accounting at ACP Facility.

Sample Matrix & Measurement Method	Uncertainty Component (% rel.Std. Uncertainty)			
	So-called Random	So-called System	Sampling	Reference & Notes
DA : Spent Fuel Powder	0.2	0.2	10.0	• U & Pu by IDMS at Hot Cell
NDA : Spent Fuel Powder	4	1.5		• Pu mass by HLNC for MOX
NDA : Hulls & Wastes	10	5		• Pu mass by HLNC for MOX Scrap
DA : U-Metal	0.2	0.2	10.0	• U & Pu by IDMS at Hot Cell
NDA : Dirty Scrap	10	3		• Pu mass by HLNC for MOX Scrap
NDA : U-Metal	4	1.5		• Pu mass by HLNC for MOX
Weigh	0.05	0.05		• Electronic Balance

standpoint of facility accounting that all items in inventory be associated with measured values. Such measured values should be obtained in a way compatible with efficient operation. The destructive assay (DA) measurements for plutonium concentration are made on a batch basis. It is unnecessary, time consuming, and costly to obtain a sample from each individual container of powder. Instead, samples are drawn from containers deemed representative of other containers in a batch.

The facility's material control and accountability methods propagate all measurement and sampling uncertainties to give a standard error. As shown in Table 2, the measurement methods used for the material accounting are assumed to have various uncertainties based on the ITV 2000 [10]. The measurement precisions and accuracies reflected in the table by the random and systematic uncertainties, respectively, are values achieved in the analysis of materials of nuclear grade or similar chemical impurity. They include the contributions of all uncertainties occurring after sampling. Using these assumptions and uncertainty values, the result for the σ_{MUF} was estimated as 1.602 kg of elemental plutonium, assuming no data falsification. The corresponding limit of error value for MUF is 3.204 kg of plutonium. This result suggests that it would be possible to meet typical IAEA detection goals for campaigns having 3 months or fewer.

It has been pointed out that the primary role of inspection from an accounting viewpoint is to install confidence in the reported MUF and its variance. In performing this function, the so-called D-statistics, or the difference statistic, is of prime importance. The quantity D is an estimate of this bias in the facility MUF. In actuality, it estimates a relative bias between the facility and the inspection agency, which is interpreted as a bias in the facility MUF when the assumption is made that the agency inspection measurements are unbiased.

For the D-statistics estimation in the facility, it was assumed that only one type of NDA measurement per item is used for verification accounting, with no destructive samples and no attributes measurements. For the inspection plan developed for the conceptual ACP facility, σ_D was estimated as 2.5 kg of plutonium. Thus σ_D is roughly 2.8% of the total plutonium handled during MB period. The largest single contributor to σ_D involves PWR powder measurement. In practice, the value of D will not equal zero because of measurement errors on the parts of the facility (for declared values) and the inspectorate (for verification values). In most cases, σ_D greatly exceed σ_{MUF} because the inspectorate's accounting is based on poorer quality measurements (e.g., NDA vs. DA) of fewer items. It is necessary to compare D to a limit, based on propagation of the uncertainties

involved, to evaluate the possibility of data falsification. From the D statistics results, it could be concluded that the sensitivity of the verification for conceptual ACP facility is very good because the inspection plan affords good protection against gross falsification and σ_D small relative to 1 SQ (8 kg) of plutonium. This calculation is a preliminary estimate that is expected to be modified as more information becomes available about measurement performance.

SUMMARY

A preliminary study on the safeguardability of a pilot-scale ACP facility was performed. As a result of the study, our conceptualization of facility features and material flows across the ACP facility lead us to conclude that a safeguards system could be designed to meet the IAEA's detection goals and to provide an independent verification scheme. During and following the selection of an ACP option for engineering demonstration, parallel efforts will be directed at developing systems for material accounting, measurements, containment and surveillance, and verification of the flow and inventories of materials at the ACP facility. As we get information on measurements and verification approaches that is more reliable, these data and calculations can be modified.

ACKNOWLEDGEMENT

This work was sponsored by the US Department of Energy, Office of International Safeguards, and the Korea Ministry of Science & Technology, Nuclear R&D Program.

REFERENCES

- [1] RadWaste Management Center, <http://www.4energy.co.kr/know/know1.html>
- [2] Ackerman, J. P., Johnson, T. R., and Laidler, J.J., "Waste Removal in Pyrochemical Fuel Processing for the Integral Fast Reactor," Actinide Processing: Methods and Material, TMS Publications, Warrendale, 1994.
- [3] K. Budlong-Sylvester, K. E. Thomas, and J. F. Pilat, "Proliferation Resistance for AFCI," LA-UR-03-0501, LANL (2003).
- [4] I. S. Kim, et al., "Characteristics of Reduced Metal from Spent Oxide Fuel by Lithium," Journal of the Korean Nuclear Society, Vol. 35, No. 4, 309-317, 2003.
- [5] IAEA, "INFCE Working Group 4 Report," INFCE/PC/2/4, IAEA, Vienna (1980).
- [6] W. H. Hannum, et al., "Nonproliferation and Safeguards Aspects of the IFR," Progress in Nuclear Energy, Vol. 31, No.1/2, 203-217, 1997.
- [7] H. D. Kim, et al., "A Preliminary Study on the Safeguardability of Advanced Spent Fuel Conditioning Process," INMM 44th Annual Meeting, Phoenix (2003).
- [8] T. K. Li, et al., "Safeguardability of Advanced Spent Fuel Conditioning Process," LA-CP-04-0132, LANL (2003).
- [9] D. Vaden, R. W. Benedict, K. M. Goff, "Material Accountancy in an electrometallurgical Fuel Conditioning Facility," Argonne National Laboratory, CONF-9606116-40 (1996).
- [10] Aigner, H., et al., "International Target Values 2000 for Measurement Uncertainties in Safeguarding Nuclear Materials," STR-327, IAEA, Vienna (2001).